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SINGLE-STAGE PYROMETALLURGICAL PROCESSING OF RADIOACTIVE SCRAP METAL

Gudim Yu. A.[1], Golubev A. A.[1], Grachev V. A.[2]

[1]**Industrial company "Metals Technology", Chelyabinsk, Russian Federation**

[2]**A.N. Frumkin Institute of Physical Chemistry and Electrochemistry, Center for Global Ecology of Lomonosov Moscow State University, Moscow, Russian Federation**

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The authors proposed a method for a single-stage pyrometallurgical processing of radioactive scrap metal using an experimental facility of the MAGMA melting unit. Feasibility of implementing the proposed technological scheme in semi-industrial conditions and producing decontaminated metal suitable for economic use without restrictions has been confirmed. The methods of trapping and safe conditioning of small amounts of secondary solid radioactive wastes were justified and verified. A pilot project for a single-stage decontamination of radioactive scrap metal in the MAGMA-3 melting unit has been carried out.

Keywords: *metallic radioactive waste (MRW), pyrometallurgic decontamination, single-stage process, melting facility, acidic slag.*

Introduction

Intense economic and military activities of mankind had generated enormous amounts of radioactive waste (RW), with a considerable part being radioactive scrap metal or metallic radioactive waste (MRW).

RW needs to be removed from the human habitat by disposal (for high-level waste — HLW) or by reprocessing for subsequent reuse (for all other types of waste).

Background

It is advisable to recycle radioactive scrap metal. The most effective method of MRW decontamination is pyrometallurgic decontamination. Only two-stage or multi-stage processes of MRW decontamination have been used up to this moment. All these processes applied preliminary low-temperature removal of surface contamination by various means (physical-mechanical, physical-chemical, chemical). Pyrometallurgic method was used only for the final treatment of partially decontaminated metals (melting in induction and arc electric furnaces) [1—12]. A. A. Vertman was the first to propose single-stage pyrometallurgic decontamination of radioactive scrap metal [12]. He also proposed general solutions for the design of the appropriate melting facility.

The idea of single-stage MRW decontamination proposed by A. A.Vertman is a promising one and can be implemented. However, the melting facility design he suggested cannot be used for radioactive scrap decontamination due to some design deficiencies.

Traditional induction and arc electric furnaces are unsuitable for implementation of the concept of single stage MRW decontamination due to some design features.

Relevance of the topic

The multi-stage methods of radioactive scrap metal decontamination are cumbersome and have relatively low capacity to be considered a solution for the problem of already accumulated and newly generated MRW. Therefore, there is a relevant problem of development of the concept and implementation of single-stage pyrometallurgic MRW processing in melting facilities having fairly high capacity and effective process of MRW decontamination.

The authors of this paper have developed a concept of single-stage pyrometallurgic radioactive scrap metal processing in melting facility with heat removal from the surface by liquid metal coolant. Airtight fuel-oxygen skull melting facility of continuous action "MAGMA-3" was suggested for radioactive scrap metal decontamination. The top part of the facility is cooled by liquid metal sodium coolant and is not lined by refractory materials, and the lower part of the casing is lined by refractory materials producing metal pool for molten liquid metal [15].

Melting facility "MAGMA-3" was designed as a multi-functional facility capable of processing ores of various metals, as well as process and general waste. "MAGMA-3" facility was developed by LLC Industrial Company "Metal Technology" and LLC STF "Akont" (Chelyabinsk).

"MAGMA-3" facility is capable of competing with electric furnaces widely used for melting metal charges (scrap metal, metallized raw materials) [13].

The concept of single-stage pyrometallurgic radioactive scrap metal processing in liquid metal cooled melting facility was best described in [14]. In this paper not only doubtless advantages of the suggested MRW decontamination process were described, but also the problems were identified, which needed to be resolved at a small experimental bench of the melting facility in order to greatly reduce the time of construction and industrial implementation of the line for pyrometallurgic decontamination of MRW. These problems included:

- specification of the technology of sodium cooling;
- assessment of the safety of sodium-cooled melting facility in emergency situations;
- specification of behaviour of most frequent radionuclide contaminants found in MRW in process of radioactive scrap metal processing;
- assessment of the capabilities for trapping and conditioning of secondary solid radioactive waste;
- development of a method for effective conditioning of secondary radioactive dust and slag generated in pyrometallurgic MRW processing;
- development of a rational design of a container for collection, transportation and disposal of generated secondary SRW;
- design of pilot line for pyrometallurgic processing (decontamination) of radioactive scrap metal.

The goal of the current work was to investigate and resolve these problems.

Methods used in theoretical studies and experiments

MRW contaminants behaviour in process of pyrometallurgic processing was studied at a specially constructed experimental melting facility (EMF) at the laboratories of JSC "SSC RF IPPE". Diagram of EMF is shown in Fig. 1. EMF consisted of three modules:
1) pyrometallurgic processing module (Fig.2);
2) liquid metal cooling circuit module (Fig.3);
3) gas trapping and purification system module.

Induction furnace was used as the pyrometallurgic processing module. Specially prepared MRW specimen (5 pieces) had generally surface radioactive contamination. The specimen and acidic slag (tailings of JSC "Ufaley-nickel") were placed inside a refractory crucible in the induction oven. Metal and slag were melted and mixed for 30 minutes, until the temperature of the metal (stainless steel 10X18H10) reached 1600 °C. The oven was then turned off, the crucible with metal and slag was extracted and cooled. The obtained metal and slag were analyzed with respect to radioactive contamination. Dust from the gas purification system was also analyzed.

The investigation results and solutions for the above problems are given below.

1 Specification of the technology of melting facility surface cooling by sodium

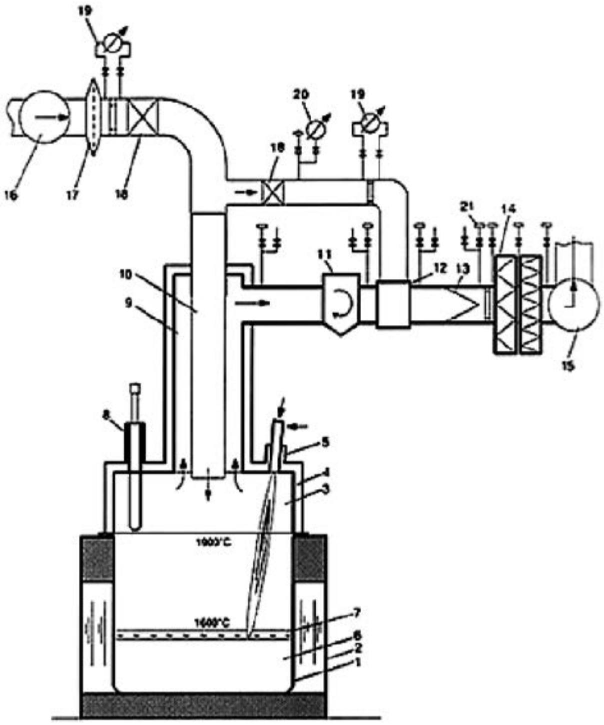
Analysis of the design and operation modes of the melting facility was carried out, including specifying the source of admixtures in the liquid sodium. Methods to reduce the amount of admixtures ingress to the liquid sodium were suggested.

The following was justified based on the results obtained: possibility of development of sodium purification systems; selection of equipment to maintain the parameters of the required sodium cooling process; recommendations for inclusion in the design of the melting facility. Sodium purification procedure without using sodium flow rate agitator and relevant system parameters was justified in calculations. Recommendations were suggested for the inclusion of the sodium purification system without the flow rate agitator, and inclusion of electric chemical analyzer of oxygen content in sodium in the cooling system of the melting facility.

The proposed recommendations were used to develop the new design of liquid sodium purification system, which did not include sodium flow rate agitator, and which could be mounted directly inside the casing cavity of the melting facility "MAGMA-3".

2 Assessment of the safety of sodium-cooled melting facility in emergency situations

The most probable and hazardous emergency situation in operation of a liquid sodium-cooled melting facility is a failure of the cooling circuit with a leak of liquid sodium to the process section of the



1 — ceramic crucible; 2 — melting assembly; 3 — incineration area; 4 — sodium cooled steel lid of the melting assembly; 5 — gas burner to increase the temperature of the gas phase; 6 — MRW; 7 — slag; 8 — pyrometer in a ceramic shell; 9 — mixer chamber; 10 — ceramic tube of the mixer chamber; 11 — precipitator; 12 — mixer chamber; 13 — sock filter; 14 — aerosol filter; 15 — gas blower; 16 — air blower; 17 — filter; 18 — valve; 19 — flowmeter; 20 — compound gauge; 21 — analytical filter

Fig. 1. EMF diagram

Fig. 2. "Hot" crucible after the experiment

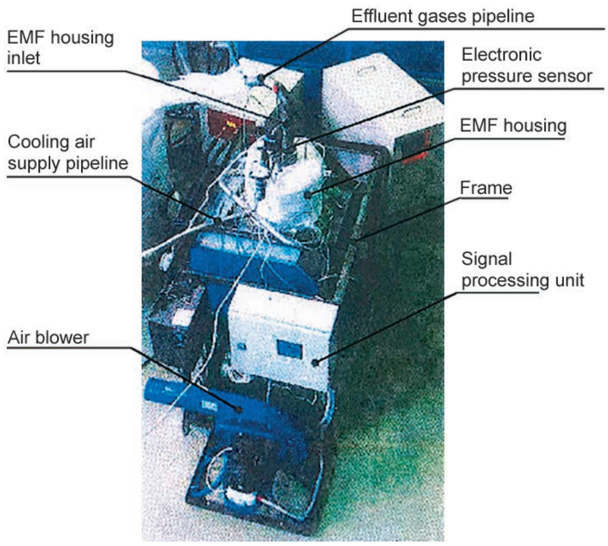


Fig. 3. Liquid metal cooling circuit module

melting chamber or to the air-filled secondary cooling circuit of the melting chamber.

Calculation methods were used to assess the safety of the melting facility in case of an emergency leak of sodium. Results of reference calculations of the experiment demonstrated that:

- sodium released to the melting area of the facility was immediately oxidized to sodium oxide Na_2O;
- the majority of generated alkali oxide Na_2O (97%) was absorbed by acidic melting slag forming complex compound $2Na_2O{\cdot}CaO{\cdot}3SiO_2$;
- the remaining 3% of Na_2O were released to the gas phase in the form of sodium salts (predominantly $NaPO_3$), removed from the operational area, and, after gas cooling, were mainly transformed to airborne phase as $NaNO_3$;
- for emergency sodium leaks with flow rate of 60 kg/h the slag mass increased by approximately 6%, the mass of effluent gases remained virtually the same;
- temperature of effluent gases increased slightly due to heat generating sodium oxidation reactions inside the melting facility, however the extent of this increase did not affect the operation of the facility;
- sodium released to the air-filled secondary cooling circuit of the melting chamber was oxidized to Na_2O oxide and was removed to the gas purification system.

Thus, the calculation results demonstrated that emergency leaks of sodium from the primary melting facility cooling circuit did not lead to immediate failure of the melting facility and evolution of design-basis accident scenarios.

Control system of "MAGMA-3" melting facility contains devices and subsystems for detection of sodium leaks and shut down of the facility at the early stage of emergency sodium leak. This further reduces the possibility of melting facility failure.

3 Specification of behaviour of most frequent radionuclide contaminants found in MRW in process of pyrometallurgic processing

Averaged results of the experiments are shown in table 1.

Data of Table 1 demonstrate that in process of heating and melting radionuclides contaminating the surface of MRW specimen have partially been transferred and bound in the acidic slag, but predominantly have evaporated, transferred to the gas phase and subsequently transferred to airborne phase in process of gas cooling.

Table 1. Results of spectrometric analysis of melting products before and after metal decontamination

Isotope	MRW specific activity, Bq/g	Specific Activity of decontaminated metal, Bq/g	Specific activity, Bq/g		Decontamination factor
			Slag	Dust	
Cs-137	216.0	⩽ 0.1	20.0	196.0	⩾ 2160
Am-241	68.6	⩽ 0.1	7.0	61.6	⩾ 686
U-235	20.0	⩽ 0.1	2.0	18.0	⩾ 200
*Co-60	110.0	1.0	50.0	60.0	110.0
Total	414.6	⩽ 1.0	79.0	317.6	⩾ 3156

* Note: Co-60 in surface deposits.

The described experiments confirmed long-term operational capability of liquid metal cooling circuit for gas phase temperature in the EMF up to 1900 °C.

The concept of dry purification of effluent gases suggested in the conception was verified using the gas trapping and purification system module. The extent of effluent gas purification obtained in the experiments was 99.99 %. Sock and aerosol filters were used in the gas purification system.

The gases were cooled to temperatures below 150 °C and above 60 °C (dew point) prior to purification. Results of the experiments confirmed the capabilities of the suggested dry system of gas purification from dust and aerosols, which does not lead to generation of liquid RW.

Thus, the technological procedure of single stage pyrometallurgic processing (decontamination) of radioactive scrap metal in a liquid metal-cooled melting facility tested in EMF experiments at JSC "SSC RF IPPE" has confirmed the possibility of MRW decontamination using industrial pilot facility "MAGMA-3" and the capabilities of liquid metal cooling, and gas purification systems suggested in the conception.

4 Verification of the capability of reliable conditioning of secondary SRW generated in radioactive scrap metal decontamination

The products of pyrometallurgic decontamination in addition to "clean" metal include secondary solid radioactive waste: final slag (up to 6 % of mass of reprocessed MRW) and dust trapped in gas purification system (up to 1 % of reprocessed MRW). As the mass of produced secondary SRW is substantially less than the mass of reprocessed MRW, the radioactivity level of these SRW is higher than the radioactivity level of MRW being reprocessed.

Therefore, there remains the problem of conditioning the SRW generated in the process for disposal or long-term storage.

The proposed conception of MRW reprocessing includes decontamination of MRW in the presence of acidic slag. The produced slags remain acidic (have low base properties (CaO)/(SiO_2) ⩽ 0.8). Such slags are not spilled in storage and cannot be washed out by water or weakly acidic solutions.

Acidic slags similar to slags produced in MRW decontamination (although not contaminated) were tested against the standard method at the central laboratory of FSUE "PA "Mayak" and demonstrated compliance with RW acceptance criteria for storage and disposal.

Actual radionuclide-containing end products of MRW decontamination process in the EMF were tested against the acceptance criteria in the conditions of JSC "SSC RF IPPE" (Table 2).

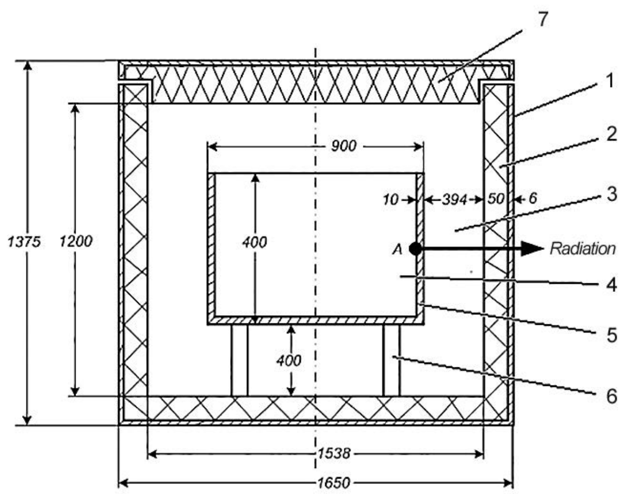
Table 2. Results of tests of end slag of EMF decontamination process against the acceptance criteria for conditioning and long-term storage

Specimen #	Leached sample activity for Cs-137			Cs-137 leaching rate for 7 day, g/cm²·day
	1 day	3 days	7 days	
1	$9.5 \cdot 10^3$	$1.39 \cdot 10^3$	$4.51 \cdot 10^2$	$4.34 \cdot 10^{-4}$
2	$1.36 \cdot 10^4$	$8.47 \cdot 10^3$	$1.28 \cdot 10^3$	$8.17 \cdot 10^{-4}$

The slag samples were considered to have passed the test if radionuclide leaching rate for Cs-137 was less than $1 \cdot 10^{-3}$g/cm·day [GOST R 50926, NP-019-15].

Thus, the end product acidic slags obtained in MRW decontamination process could be considered to be a matrix for radionuclide immobilization suitable for long-term storage and disposal.

As the mass of dust trapped by the gas purification system in process of MRW decontamination is 100 times less than the MRW mass, its level of contamination is 2 orders of magnitude higher than that of MRW. As a result, such dust is considered



1 — steel shell of the large container; 2 — acidic liner filling; 3 — acidic end slag; 4 — radioactive dust; 5 — steel shell of small container; 6 — small container support; 7 — metallic lid of the large container with refractory lining

Fig. 4. Diagram of emplacement of secondary SRW in the disposal container

to be intermediate-level waste (ILW) and there is a problem of its reliable conditioning and disposal.

The following procedure and method of conditioning radioactive dust captured by dust purification system was suggested in the conception of single-stage pyrometallurgic processing of MRW.

Standard dimension metallic container lined on the inside by a layer of acidic refractory filling 50 mm thick was suggested for storage of radioactive slag and dust.

Such a container is placed on a self-propelled rail trolley and is moved under the gutter for drain of slag from the melting facility. Dust from gas purification system is loaded to not lined metallic container of smaller dimensions than the large container used for RW disposal. Then the dust-containing container with the closed lid is placed inside a large container placed under the slag drain tray and the remaining free volume is filled with acidic end slag of the MRW decontamination process (Fig. 4). Once the slag is cooled and solidified, the large container is closed by a lined lid, sealed and sent for temporary storage or disposal.

The layer of solidified slag in the large container greatly reduces the level of γ-radiation from the external surface of the container and allows safe transportation of the container to the disposal facility. The solid mass of solidified acidic slag prevents RW release even in emergency situations.

Working design project of pilot demonstration facility for pyrometallurgic radioactive scrap metal decontamination in "MAGMA-3" melting facility was developed in accordance with the conception of single-stage pyrometallurgic radioactive scrap metal processing in liquid metal-cooled melting facility and using the results of its testing and verification. The design of the system complies to the requirements to radiation-hazardous facilities. The design is specific for the JSC 'SSC RF IPPE" site, but can be adapted to other sites with minor changes. Table 3 lists comparative parameters of "Magma-3" project and the technologies of radioactive scrap metal decontamination which are currently used or have been suggested.

The data presented demonstrate that the suggested conception of single-stage pyrometallurgic radioactive scrap metal processing in a liquid metal-cooled melting facility has a number of significant advantaged over the available MRW decontamination technologies.

Table 3. Comparison of the parameters of the project and available MRW decontamination technologies

Parameter	"MAGMA" Project	Existing technologies
Process	Single-stage	Two and more stages
Economic effectiveness	Yes	No
Capability to reprocess various types of waste	Yes	No
Energy consumption*	Low (2100 kW h/t)	High (3000 kW h/t)
Capacity for reprocessed MRW	High (up to 7000 m^3/year)	Low (up to 2500 m^3/year)
The need for preliminary decontamination and fragmentation	No	Yes
Level of activity of waste accepted for reprocessing	LLW, ILW	LLW
Type of melting facility	Fuel-oxygen, skull, airtight	Electric induction, arc and vault furnaces with refractory footage
Generation of secondary decontamination radioactive waste	Slag, dust	Slag, refractory materials, dust, liquid RW

* Note: calculated with respect to primary energy

Conclusion

Trial operation of the technology of single-stage pyrometallurgical processing of radioactive scrap metal in an experimental melting facility has confirmed the viability of implementation of the suggested procedure in half-industrial conditions with production of decontaminated metal suitable for economic use without restrictions. The methods of trapping and safe conditioning of small amounts of secondary solid radioactive wastes were justified and verified. The results have been used in the developed design of pilot demonstration system for single-stage pyrometallurgic radioactive scrap metal processing in liquid metal-cooled melting facility.

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Information about the author

Golubev Anatoly Anatolievich, Industrial Company Metals Technology, Deputy General Director, e-mail: a.golubev@metalstech.ru.

Grachev Vladimir Alexandrovich, Corresponding Member of the Russian Academy of Sciences, professor, Doctor of Technical Sciences, A. N. Frumkin Institute of Physical Chemistry and Electrochemistry of Russian Academy of Sciences, Center for Global Ecology of Lomonosov Moscow State University, e-mail: eco.moskvitina@mail.ru.

Gudim Yury Alexandrovich, Doctor of Technical Sciences, Deputy General Director for Research, Industrial Company Metals Technology, e-mail: mail@metalstech.ru.

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